

Development of High Coercive Powder From the Nd-Fe-B Sintered Magnet Scrap

A. S. Kim, D. H. Kim, S. Namkung, T. S. Jang, D. H. Lee, H. W. Kwon, and D. H. Hwang

Abstract—There have been many studies to recycle abandoned Nd-Fe-B magnets. Of these, some studied the conversion of sintered Nd-Fe-B magnet scrap to bonded magnet powder. This conversion, however, has never been used in production yet. The present study aims to develop a highly coercive magnet powder for bonded magnets from Nd-Fe-B sintered magnet scrap. In general, the magnetic property of the powders is dramatically lost through crushing of the sintered magnet because of microstructural damage. The magnetic property of the powders is partly recovered by a heat-treatment due to the healing of microstructural damages. The coercivity of the powders could be fully restored by heat-treatment with a surface modifier such as DyF_3 . Surface-morphology change and microstructural uniformity may lower the nucleation sites of reversed domains, which may be responsible for recovery of demagnetization loop squareness and an increase in H_c . An anisotropic powder obtained from the Nd-Fe-B magnet scrap exhibits $B_r = 11.6 \text{ kG}$, $H_c = 13.3 \text{ kOe}$, and $(BH)_{\text{max}} = 31.1 \text{ MGOe}$.

Index Terms—Coercivity, magnet scrap, rare earth magnet (Nd-Fe-B), recycling.

I. INTRODUCTION

SINCE its introduction in 1984 [1], [2], the Nd-Fe-B magnet has revolutionized electronic and electric products through volume miniaturization and because of its very strong magnetic properties. Because of its high performance, its application area has rapidly expanded. The advance of industry makes our life more convenient. However, it has brought about environmental problems as its use has increased in many consumer products. The products using these magnets should be recycled in order to preserve the environment. There have been many studies on how to recycle used Nd-Fe-B magnets. One [3] study examined the separation and extraction of rare earth (R) from the Nd-Fe-B magnet scrap or sludge. Another [4] studied re-melting the magnet scrap, and yet another [5], [6] examined the conversion of sintered Nd-Fe-B magnet scrap to bonded magnet powders. Rivoirard *et al.* [5] reported that the hydrogen decrepitation and desorption (HD) process applied to bulk wastes of sintered magnets produced anisotropic Nd-Fe-B powders with increased performance and reduced costs. Although the magnetic properties are somewhat increased by the HD process, the produced powder is not stable at elevated temperatures and lose coercivity. As a result, the powder produced by HD has never been used in

bonded magnet production yet. The reason why HD powder has not been developed for bonded magnet use has more to do with commercial viability, as a well-established adequate product is available.

On the other hand, Croat *et al.* [2] developed a highly coercive nano-structured isotropic Nd-Fe-B powder by a melt spinning process. The powder has been well adopted in bonded magnet production and has contributed to the development of many new applications. However, its application has been limited partly because of low magnetic properties due to the isotropic nature, and partly because of relatively high price of the powder. Therefore, there is a need to develop an anisotropic Nd-Fe-B powder with high magnetic properties. Takeshita *et al.* [7] developed a highly coercive anisotropic Nd-Fe-B powder by the HDDR (hydrogen disproportionation, desorption, and recombination) process. Because of anisotropic nature, the powder exhibits much higher magnetic properties than the isotropic powder ($\sim 40 \text{ MGOe}$ versus 15 MGOe). Although the HDDR powder has very high magnetic properties, its application has not been well developed because of high production cost. In light of this need, a stable, anisotropic high performance, and low cost Nd-Fe-B powder should be developed. In addition, an efficient recycling process for the abandoned magnets should be developed. To meet these requirements, we attempted to develop an anisotropic Nd-Fe-B powder from the abandoned (very cheap) Nd-Fe-B sintered magnet scrap.

II. EXPERIMENT

A sintered magnet scrap of $\text{Nd}_{14}\text{Fe}_{61}\text{B}_6$ was used in this study. The bulk magnet was crushed by a mechanical method (MC) or by hydrogen decrepitation (HD) method. The crushed powders were sieved under -150μ . The magnetic properties of a bulk sintered magnet (3 mm cube), HD powders (-150μ) and MC powders (-150μ) were measured with a vibrating sample magnetometer (DOEI VSM-5-19) at room temperature. The 100 grams each of HD and MC powders were loaded in separate stainless steel boats and heat treated in a vacuum furnace under vacuum at different temperatures from 500°C to 1100°C . The same amounts of HD and MC powders mixed with some additives including 5% dysprosium fluoride (DyF_3) powder, respectively, were also heat-treated under the same condition. The magnetic properties of powders heat-treated were measured with a VSM. The powders were aged at temperatures between 150°C and 300°C for 2 h in an oven. The coercivities of aged powders were measured and plotted against aging temperature. The surface and cross section of powders of as-crushed, heat-treated, heat-treated with DyF_3 mixture were examined under a scanning electron

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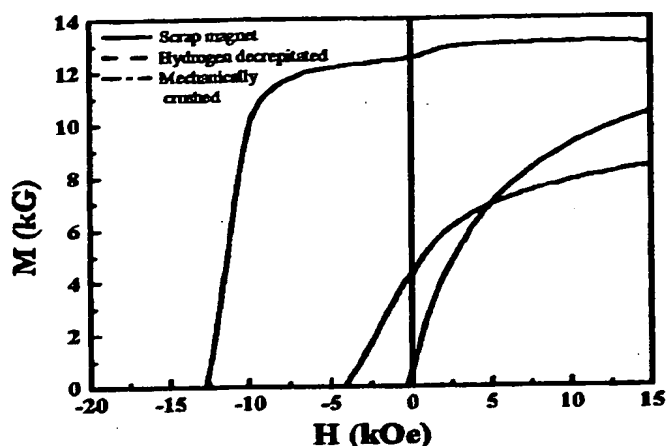


Fig. 1. Demagnetization curves of a bulk Nd-Fe-B sintered magnet scrap, mechanically crushed magnet powders (MC), and hydrogen decrepitated and crushed magnet powders (HD).

microscope (Hitachi N3000 SEM) with an energy dispersive X-ray spectrometer (EDAX EDS).

III. RESULTS AND DISCUSSION

The bulk Nd-Fe-B sintered magnet (or its scrap) usually exhibits high magnetic properties, particularly high coercivity (iH_c) as shown in Fig. 1. However, when it is crushed into powder through mechanical crushing (MC) or hydrogen decrepitation (HD), it loses its magnetic properties dramatically as shown in Fig. 1. Therefore, the as-crushed powders (either MC or HD) cannot be used as permanent magnet powders. In order to make useable magnet powders, the powders should have a high coercivity. The coercivity (iH_c) at the operating temperature should be greater than half of the residual magnetization (B_r) value of the magnet in order to assure a high energy-product. To obtain high coercivity, the HD powders were heat-treated at various temperatures under vacuum for hydrogen desorption as suggested by Riviorard *et al.* [5], and the MC powders were also heat-treated under the same condition. As reported, the heat-treatment increases the magnetic properties of HD powders as well as MC powders as shown in Fig. 2. The demagnetization loop-squariness was substantially improved. The heat-treated HD and MC powders obtain the maximum (iH_c) at 1000 °C heat-treatment. However, these values (6–8 kOe) are still far lower than that (~13 kOe) of the bulk magnet shown in Fig. 1. To examine if these powders can be used for bonded magnets, the temperature stability of the powders was examined by measuring the coercivity as a function of aging temperature. It is noted that the curing temperature for the compaction bonded magnet ranges from 150 °C to 200 °C and the injection molding temperature ranges from 200 °C to 300 °C. Therefore, the powder should be stable at these temperature ranges. As shown in Fig. 3, the coercivities of heat-treated HD and MC powders substantially decrease as the aging temperature increases. In other words, the coercivities of heat-treated HD and MC powders after aging are too low to be used for bonded magnets. Therefore, it is necessary to fully recover the coercivity of the sintered magnet.

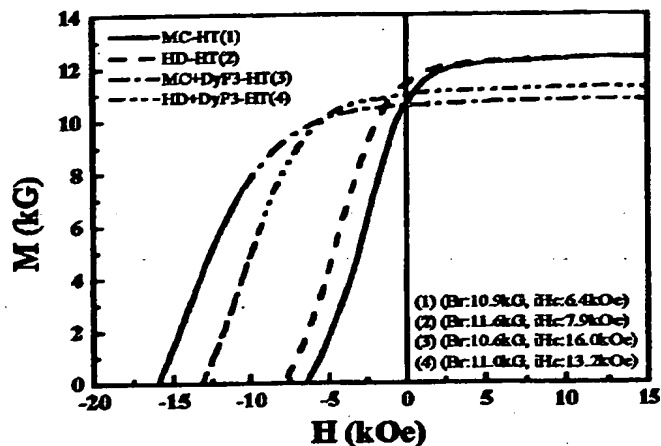


Fig. 2. Demagnetization curves of MC and HD powders heat-treated at 1000 °C for 2 h and those with DyF₃ additive.

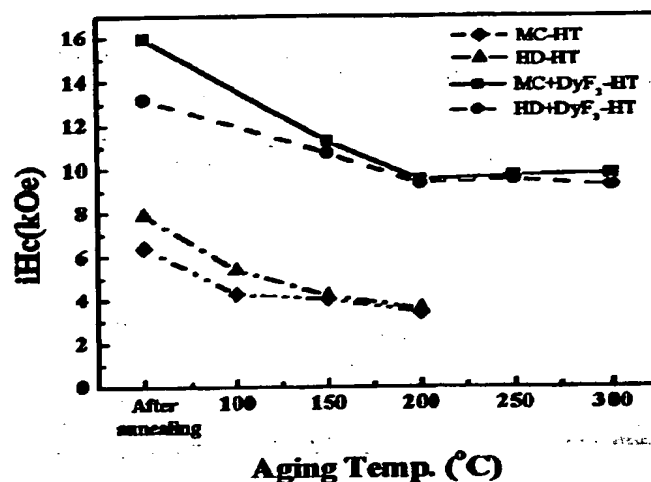


Fig. 3. Variations of iH_c of MC and HD powders heat-treated with and without DyF₃ additive as a function of aging temperature.

To fully recover the coercivity, the particle surface was modified by adding some surface modifiers before heat-treatment. Various additives of rare earth compounds were added to the as-crushed powders (both HD and MC), and well mixed before heat-treatment at 1000 °C. It was found that additions of rare earth fluorides (particularly DyF₃) substantially increase the (iH_c) from that of the heat-treated powders without additive. Fig. 2 shows the demagnetization curves of heat-treated MC and HD powders with 5% DyF₃ additions, respectively. It is noticed that the coercivities of DyF₃ added powders are much further increased from the as heat-treated powders without additive. Comparing with the demagnetization curve of the bulk sintered magnet shown in Fig. 1, those of the heat-treated powders with DyF₃ additive are very similar. It can therefore be said that the coercivity (iH_c) is fully recovered by the heat-treatment with DyF₃ additive. The thermal stabilities of the heat-treated powders with DyF₃ additive were examined at different aging temperatures as shown in Fig. 3. The coercivities of both MC and HD powders heat-treated with DyF₃ additive are somewhat

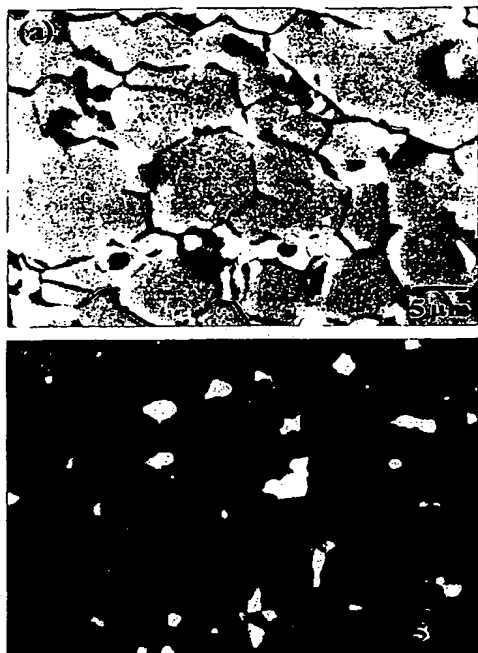


Fig. 4. SEM micrographs showing cross sections of as-crushed and heat-treated powders.



Fig. 5. SEM micrograph showing cross section of a powder heat-treated with DyF_3 additive.

decreased by aging from the as heat-treated condition, but maintain about 9–10 kOe regardless of the aging temperatures. These coercivities (9–10 kOe) are much higher than those of the aged powders without DyF_3 additive, and are good enough ($>1/2D_r$) to make high performance bonded magnets. As a result, we have developed a highly coercive powder for bonded magnets from the Nd-Fe-B sintered magnet scrap by heat-treatment of crushed powders with a surface modifier of dysprosium fluoride.

The (iH_c) increase of the heat-treated powders was studied by examining the cross section of the particles under the SEM. The particles were mounted in an epoxy resin, ground and polished with 0.5 μ diamond paste. It is apparent that the HD crushed powder exhibits grain boundary cracks, while the heat treated powder exhibits a sound microstructure without cracks as shown in Fig. 4(a) and (b). The healing of grain boundary cracks by heat-treatment and microstructural uniformity may

be responsible for the improvement of demagnetization loop squareness and increase of (iH_c). Further increase in (iH_c) of the DyF_3 added powder was also investigated by examining the microstructure under the SEM with EDS. Fig. 5 exhibits a SEM micrograph of the cross-section of a particle heat-treated with DyF_3 additive. The R-rich grain boundary phases are partly discolored from white to gray, and the surface of the particle was smoothened. The EDS analysis on gray Nd-rich phase exhibits high Nd and F peaks, while the analysis on white Nd-rich phase exhibits mainly a Nd peak. The EDS analysis on the particle surface exhibits high Dy, Nd and Fe peaks. These indicate that DyF_3 additive may decompose to Dy and F on the particle surface during the heat-treatment, where the F diffuses in through the grain boundary and enhances microstructural soundness and uniformity, and the Dy diffuses in through the particle surface and is enriched at the near surface of the powder particle. The surface of the particle is also smoothened. The surface-morphology change and uniform microstructure may lower the nucleation sites of the reversed domains, which may be responsible for the recovery of the coercivity of the powders. This result is similar to the result obtained from coating a thin Nd-Fe-B magnet with Dy metal by radio frequency sputtering and subsequent heat treatment, which improved the coercivity about twice as much as that of the original thin magnet [8].

A low cost, highly coercive and stable anisotropic Nd-Fe-B magnet powder was produced from the cheap and abandoned Nd-Fe-B magnet scrap. In other words, the abandoned Nd-Fe-B magnet scrap can be recycled into a valuable bonded magnet powder.

IV. CONCLUSION

A highly coercive magnet powder for bonded magnets was developed from the Nd-Fe-B sintered magnet scrap. The coercivity of bulk Nd-Fe-B sintered magnet is dramatically lost through crushing into powders. The coercivity of the powder could be partly recovered by a heat treatment and fully restored by surface modification with some additive such as DyF_3 by thermal activation. The modification of particle surface and grain boundary may lower the nucleation sites of the reversed domains, which may be responsible for the recovery of the powder's coercivity. An anisotropic powder obtained from the Nd-Fe-B magnet scrap exhibits high coercivity and energy product. It also exhibits good thermal stability.

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